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# High-precision mass measurements for reliable nuclear astrophysics calculations

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High-precision mass measurements as performed at the Penning trap mass spectrometer ISOLTRAP at ISOLDE/CERN are an important contribution to the investigation of nuclear astrophysics processes. Accurate nuclear masses with 0.1 ppm relative mass uncertainty are required for tests of models which are used to predict mass values of nuclides far from the valley of stability. Especially for the determination of the nucleosynthesis pathways, e.g. the rp-process or the r-process, a large number of nuclear masses need to be precisely and accurately known. In addition to a radioactive ion beam facility that delivers a large variety of short-lived nuclides with sufficient yield, a sophisticated experimental setup for precise mass measurements is required. An overview of the results from the mass spectrometer ISOLTRAP is given and the limits and possibilities are described.

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## 1. Introduction

Nuclear masses play an important role in a large number of physical systems. Especially in the case of astrophysics, nuclear parameters of radionuclides far from the valley of stability are required. For the investigation of the nucleosynthesis pathways, masses with a relative uncertainty of the order of $10^{-7}$ are needed [1, 2]. However, for a large number of nuclides the present mass uncertainty is above 10 keV, and especially for short-lived nuclides the relative mass uncertainty rapidly rises the farther out the nuclides lie (see Fig. 1). Short half-lives and low production yields makes their investigation an experimental challenge.

Mass models either provide estimates on unknown masses by a global approach, or try to predict the mass values locally [3]. In order to test these mass models, accurate experimental data are required. Various experiments at radioactive ion beam facilities aim at the investigation and precise determination of the masses of short-lived nuclides [4]. The applied techniques range from traditional TOF mass spectrometers to storage rings and Penning trap mass spectrometers [5]. The latter have shown to allow mass measurements for stable ions with a relative uncertainty down to $10^{-11}$ [6] and for radionuclides of the order of or less than $10^{-8}$ [7 – 10].

The Penning trap mass spectrometer ISOLTRAP has already contributed to a number of astrophysical problems by providing accurate mass values. In the case of $^{72}$Kr, the identification of a rp-process waiting point was achieved [11] and for $^{22}$Mg a puzzling mass contradiction for the NeNa-cycle in supernovae was resolved [12]. These examples show the need for accurate mass values. In the following, recent investigations on the mass values of neutron-rich nuclides will be presented and the possibilities, challenges, and limitations discussed.

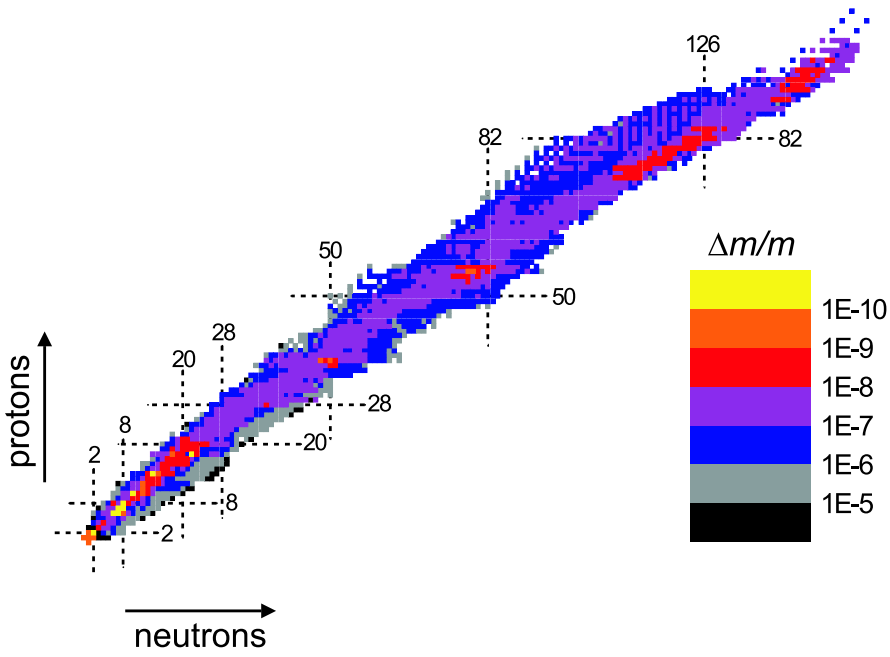


**Figure 1:** Relative mass uncertainty $\Delta m/m$ of nuclides in color code. Estimated mass values are omitted. The dashed lines indicate the neutron and proton shell closures. Data taken from [13].

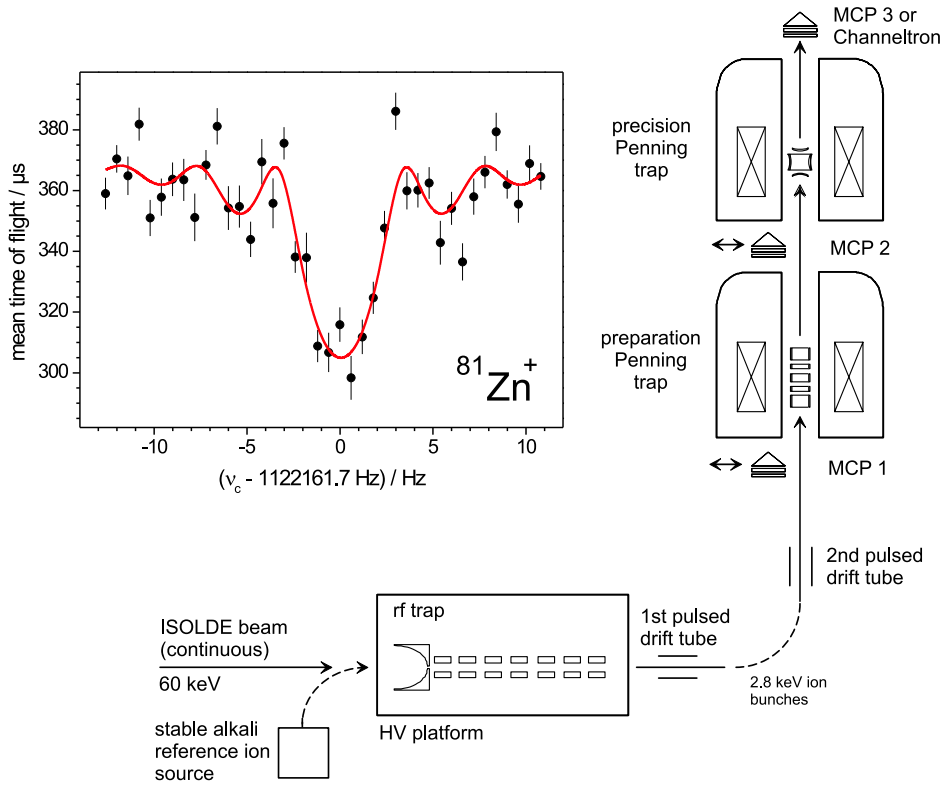


**Figure 2:** Schematic overview of the ISOLTRAP experiment. The inset shows a cyclotron resonance of $^{81}Zn^{+}$, where the solid line is a fit of the known line shape to the data points [14].

## 2. Experimental setup and procedure

The Penning trap mass spectrometer ISOLTRAP [15, 16] is the prototype experiment for the mass determination on radionuclides at radioactive ion beam facilities using Penning traps. Various additional experiments are meanwhile in operation (CPT [17], JYFLTRAP [18], LEBIT [19], SHIPTRAP [20]) or under construction (TITAN [21], MLLTRAP [22]). A schematic overview of the experimental setup is shown in Fig. 2. The radioactive ions are delivered from one of the two target stations at ISOLDE [23]. The short-lived nuclides are produced by bombarding a fixed target with a 1.4-GeV proton pulse and subsequent spallation, fragmentation, and fission reactions. The target container is heated and the radioactive particles diffuse into an ion source, where ions are formed by either surface ionization, resonant laser ionization, or a plasma. After passing separator magnets, the ions are sent as a 60-keV continuous beam to the ISOLTRAP setup.

By use of a linear radiofrequency (rf) quadrupole structure [24] filled with helium buffer gas, mounted inside a 60-kV high voltage cage, the continuous beam is stopped, accumulated, and bunched as well as cooled for further injection into the first Penning trap. This cylindrically shaped trap also employs helium buffer gas, and with a combination of a quadrupolar rf excitation and gas collisions only the radionuclide ions of interest are mass-selectively centered [25]. Thus unwanted isobaric contaminant ions can be removed with a resolving power of $R = 10^4 - 10^5$, depending on the respective mass difference and the excitation duration [26].

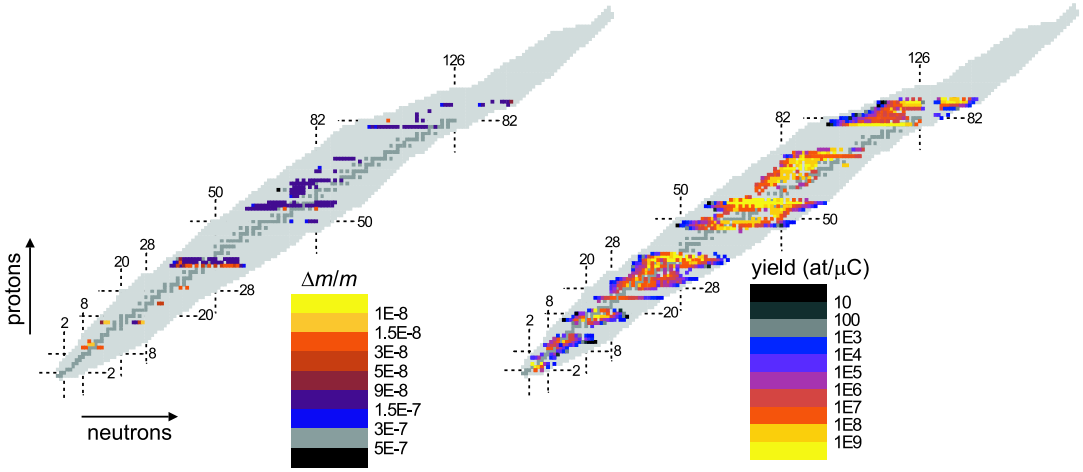


**Figure 3:** Left: Relative mass uncertainty of published mass values obtained at ISOLTRAP. Right: Maximum yields obtained at ISOLDE during operation at the Synchrotron Cyclotron (SC) and the Proton Synchrotron Booster (PSB) at CERN [27].

The cooled, isobarically pure ion bunch is transferred to the hyperbolically shaped precision Penning trap, where the cyclotron frequency $\nu_c = qB/(2\pi m)$ of the stored ions with mass $m$ and charge $q$ moving in a magnetic field $B$ is probed with a time-of-flight (TOF) cyclotron resonance detection technique [15, 28]. In order to deduce the mass of the investigated radionuclides, the magnetic field strength $B$ needs to be determined at the same level of precision, by use of a cyclotron-frequency measurement of a well-known stable nuclide ion, either delivered from the ISOLTRAP alkali ion source or the ISOLDE target. By calculating the frequency ratio, the mass ratio of the ions is obtained.

## 3. Limitations and possibilities for mass measurements at ISOLDE

### 3.1 Radionuclide half-lives and production yields

In principle, all nuclides delivered by ISOLDE can be investigated with ISOLTRAP. However, due to the preparation steps and the excitation time required for the frequency determination, the half-life is a major constraint. The nuclide with the shortest half-life investigated so far was $^{74}$Rb with $T_{1/2} \approx 65$ ms [29].

Another important factor is the production yield in the target/ion-source combination. Figure 3 (right) shows an overview of observed maximum production yields at ISOLDE. In addition to the production yield, the efficiency of transfer and stopping at ISOLTRAP has to be taken into account. The transfer from the separator magnets, through the ISOLDE central beam line to the ISOLTRAP setup can be tuned to an efficiency above 80 %. The injection and stopping as well as cooling in the RFQ buncher has an efficiency of up to 10 % [24]. Although the transfer from the buncher to the final TOF detector is thoroughly tuned before each on-line run, the total efficiency of the ISOLTRAP spectrometer is about 1-5 %, i.e. out of 100 ions released on average in the target/ion-source only one to five ions will be detected. This assumes that the half-life of the radionuclides is larger than the duration of the experimental cycle including the excitation time for

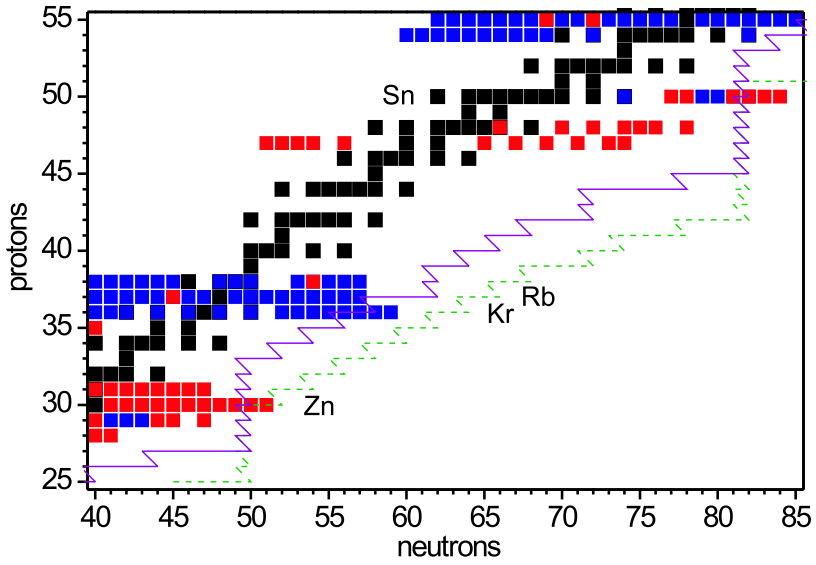


**Figure 4:** Possible pathways of the r-process (solid and dashed lines) as given in [30]. The blue squares denote nuclides for which mass values, obtained with ISOLTRAP, have been published. Red squares indicate nuclides for which data have been recorded that are currently under investigation or being published. Black squares represent stable nuclides.

the frequency determination. Ion loss due to short half-lives in the order of 100 ms and less poses another experimental challenge.

Note that some ions cannot be delivered at all, since their elemental properties prevent the diffusion out of, or ionization at the target. For example ions of refractory elements like vanadium are at the moment not available at ISOLDE. Nevertheless, new ISOLDE beams might become available in the future, if those elements are addressed by use of ion chemistry, i.e. by the production of specific molecules in the target that can easily diffuse out of the target (for example fluorides, oxides, or sulfides).

### 3.2 Isobaric and isomeric contaminants

Depending on the target material and the ion source in use, various isobaric and isomeric contaminating ions may be present in the delivered radioactive ion beam. Therefore, target development is an important issue at ISOLDE. For example, a new quartz transfer-line target was designed and built [31], which allows to freeze out alkali contaminants like rubidium ions in the transfer from the target container to the ion source. This kind of target was recently used to produce a very clean beam of neutron-rich zinc isotopes for mass measurements at ISOLTRAP.

Thus it was possible to measure the cyclotron frequencies of $^{71-81}Zn^{+}$, for which the expected contaminants, especially rubidium isotopes, were reduced to such small amounts that they could be handled and removed in the ISOLTRAP preparation trap. With a yield of 100–1000 ions per second for $^{81}Zn^{+}$, it was possible to observe on the average one ion in each experimental cycle, enough to obtain several cyclotron resonances for $^{81}Zn^{+}$ as shown in the inset of Fig. 2.

Another way to suppress or remove isobaric contaminants from the radioactive ion beam is the application of ion-molecule chemistry. Element-specific reactions can be used to produce a

| element | mass numbers | references |
|---|---|---|
| Ne | 18,19,23,24 | [32] |
| Na | 21,22 | [12] |
| Mg | 22 | [12] |
| Ar | 32–34,42–45 | [7, 33, 34] |
| Cr | 56,57 | [35] |
| Cu | 68,68m,70,70m,70n | [36, 37] |
| Kr | 72–78,80,82,84,86,88–95 | [11, 30, 34, 38] |
| Rb | 74–81,82m,83,84,86–89,90m,91–94 | [34, 39, 40] |
| Sr | 76–83,86–88,91–95 | [39 – 42] |
| Sn | 124,129,130,130m,131,132 | [42] |
| Xe | 114–123,126,136 | [43, 44] |
| Cs | 117–122,122m,123–132,134–142 | [45, 46] |
| Ba | 123–128,131,138–144 | [46, 47] |
| Ce | 132–134 | [48] |
| Pr | 133–137 | [47] |
| Nd | 130,132,134–138 | [47, 49] |
| Pm | 136–139,140m,141,143 | [47] |
| Sm | 136–141,141m,142,143 | [47] |
| Eu | 139,141,142m,143–149,151,153 | [47 – 49] |
| Dy | 148,149,154 | [47 – 49] |
| Ho | 150 | [47] |
| Tm | 165 | [48] |
| Yb | 158–164 | [48] |
| Hg | 179–188,190–197,185m,187m,189m,191m,193m,197m,200 | [50] |
| Pb | 187,187m,196,198 | [50, 51] |
| Bi | 197 | [50] |
| Po | 198 | [50] |
| At | 203 | [50] |
| Fr | 209–212,221,222,230 | [52, 53] |
| Ra | 226,229–232 | [46, 52, 53] |

**Table 1:** Unstable and stable nuclides measured with ISOLTRAP.

clean molecular sideband. As an example, the investigation of neutron-rich tin nuclides, $^{128-134}$Sn, became possible when the radionuclides of interest were shifted out of the mass range with Cs contamination, by adding an enriched sample of $^{34}$S to the target in order to form tin-sulfide molecules (SnS).

A further challenge are isomeric states with half-lives of more than 10 ms that are also produced in the target and may contaminate the radionuclide beam. While the isomeric states can usually not be resolved and isolated by the separator magnets of ISOLDE and even by the preparation trap of ISOLTRAP, the precision Penning trap allows isomeric cleaning with a resolving power

| element | mass numbers | year |
|---|---|---|
| Ne | 17 | 2004 |
| Al | 26,27 | 2006 |
| K | 35–38,43–46 | 2004 |
| Ca | 38,39 | 2006 |
| Mn | 56–57,58m,59–63 | 2003,2006 |
| Fe | 61–63 | 2006 |
| Ni | 57,60,64–69 | 2003 |
| Cu | 65–67,69,73,74,76 | 2003 |
| Zn | 71m,72–81 | 2005 |
| Ga | 63–65,69–78 | 2003 |
| Se | 70–73 | 2002 |
| Br | 72–75 | 2002 |
| Ag | 98–101,103,112,114,116,118m,120,121 | 2002,2006 |
| Cd | 114,120,124,126 | 2005,2006 |
| Sn | 127,128m,133,134 | 2004 |
| Cs | 145,147 | 2002 |
| Ba | 130 | 2003 |
| Tl | 181,183,186m,187,196 | 2002 |
| Pb | 197,208 | 2002 |
| Bi | 190–196,215,216 | 2002 |
| Fr | 203,205,229 | 2002 |
| Ra | 214 | 2002 |

**Table 2:** Unstable and stable nuclides for which data have been recorded with ISOLTRAP (year of on-line run given) and data analysis is ongoing or data will be published soon.

of the order of $R \approx 10^7$. Thus, either pure isomeric states or the ground state can be prepared as was demonstrated for $^{68}$Cu [36]. Note that if an ion source with resonant laser ionization (RILIS) is applied, narrow-band laser excitation can state-selectively populate single isomeric states or significantly suppress the unwanted ones, as demonstrated, e.g., in the case of $^{70}$Cu [37].

## 4. Recent investigations

The masses of over 300 nuclides have already been measured with ISOLTRAP. An overview is given in tables 1 and 2. For published frequency-ratio values, the corresponding relative mass uncertainties are shown in Fig. 3 (left). While in the beginning the systematic uncertainties were not known and therefore a conservative estimation of the lower limit of the relative mass uncertainty of $1 \times 10^{-7}$ was applied, the systematic investigation of the setup via the mass measurement of carbon clusters was a major improvement [54]. Carbon clusters consisting of a multiple of $^{12}$C atoms are the ideal reference masses, because the atomic mass unit is defined as 1/12 of the mass of $^{12}$C [55]. A systematic study showed the limit of the accuracy of ISOLTRAP at $\Delta m/m = 8 \times 10^{-9}$ [56]. This is added as a systematic uncertainty to the statistical uncertainty from the frequency determination.

Recent results regularly reach relative mass uncertainties of the order of $1 \times 10^{-8}$, some of those are shown in Fig. 3 (left). The experimental data of a large number of nuclides is presently being analyzed or the results are close to publication. The respective nuclides are given in table 2. These include neutron-rich zinc and tin radionuclides with the nuclides $^{80}$Zn and $^{132}$Sn, which are relevant for the study of the r-process, as illustrated in Fig. 4. In both cases the studied nuclides lie within the possible path of the r-process, and large deviations from the literature values [13] have been found. The influence on astrophysical calculations is currently investigated.

Two previous results for mass measurements at ISOLTRAP have already contributed to the study of the nucleosynthesis in stellar explosions and the rp-process. In the case of $^{22}$Mg, the former literature mass value was in conflict with results from $^{21}$Na$(p,\gamma)^{22}$Mg reaction rate measurements [57]. Indeed, the previous mass excess value -397.0(1.3) keV [58] was found to significantly deviate from the new value -399.92(27) keV [12] and a $^{21}$Na$(p,\gamma)^{22}$Mg resonance energy of 209.7(1.2) keV into the 5714-keV $2^+$ state of $^{22}$Mg was deduced. This independent determination of the resonance energy gives further input to the comparison of the observed $\gamma$ radiation from oxygen-neon nova explosions with the yield of $^{22}$Ne as expected from nova models, where $^{22}$Ne is produced in the so called NeNa-cycle [57].

The second example is the mass determination of $^{72}$Kr. This neutron-deficient nuclide, which lies close to the proton drip line, is one of the three major waiting points of the rp-process. When reaching one of these nuclides, the proton capture sequence stops, since the $(\gamma,p)$ photodisintegration rate is equal to the $(p,\gamma)$ proton capture rate and a slow $\beta^+$ decay must occur before further protons can be added. With the ISOLTRAP mass measurement the mass excess value was improved from -54110(270) keV to -53941(8) keV [11]. With an uncertainty of less than 10 keV, the new mass value allowed to contrain the effective lifetime in x-ray bursts, where at temperatures of 1-1.5 GK $^{72}$Kr remains a strong waiting point with at least 80% of its $\beta^+$ decay half-life, thus delaying the burst duration.

## 5. Technical developments at ISOLTRAP

Technical improvements of the ISOLTRAP mass spectrometer include a new ion detector [59]. The existing micro-channel-plate (MCP) detector was replaced by a channeltron detector with a conversion dynode, where secondary electrons are monitored with an efficiency close to 100 %. With this setup an increase of the total efficiency by a factor of about 3 was obtained and nuclides with a lower production yield can be studied.

Furthermore, the stability of the mass measurements is improved by stabilizing the magnetic field. It was observed that the fluctuation of the cyclotron frequency is correlated to the temperature variations in the warm bore of the superconducting magnet [60] and the pressure in the helium recovery line. Therefore, new stabilization systems have been installed which aim at stabilizing the center temperature to better than $\pm$10 mK and the helium pressure to $\pm$50 mbar.

Finally, a new technique to obtain radionuclides that are not produced at ISOLDE has been tested: the production of radionuclides by in-trap decay [61]. To this end short-lived nuclides are stored in the buffer-gas-filled preparation trap and after a sufficiently long storage period (depending on the half-life), enough daughter nuclide ions from the decay have been collected in the Penning trap for further measurements. After the usual cooling and centering procedure, they are

transferred to the precision trap for mass determination. This technique has been successfully applied to the first ISOLTRAP mass measurements of $^{61-63}$Fe isotopes, which are not available from ISOLDE. The results are currently under investigation.

## 6. Conclusion and outlook

The ISOLTRAP mass spectrometer at ISOLDE delivers accurate mass values of short-lived radionuclides. The relative mass uncertainty is currently limited to $\delta m/m = 8 \times 10^{-9}$. With the possibility to reach nuclides with half-lives well below 100 ms and production yields down to 100 ions per second, a large number of radionuclides can be investigated at ISOLDE. Together with other Penning trap mass spectrometers at radioactive ion beam facilities around the world, more and more reliable mass values will become available for astrophysical calculations and tests of mass models.

Besides target development at ISOLDE, which aims at higher yields, more accessible elements, and less contamination, the experimental setup of ISOLTRAP is constantly subject to technical development. A new ion detector for a better overall efficiency, the stabilization of the magnetic field, and new reference ion sources are recent improvements of the setup. Further development is directed at a higher accuracy in the mass determination by use of new rf excitation schemes and ions of higher charge states.

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